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PASSIVE RADON DAUGHTER DOSIMETERS

by

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Research report

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ABSTRACT

On the basis of an extensive review of the recent literature concerning passive radon daughter dosimeters, we have reached the following conclusions:

(1) Passive dosimeters for measuring radon are available and reliable.

(2) There does not presently exist an acceptable passive dosimeter for radon daughters. There is little if any hope for the development of such a device in the foreseeable future.

(3) We are pessimistic about the potential of "semi-passive dosimeters" but are less firm about stating categorically that these devices cannot be developed into a useful radon daughter dosimeter.

This report documents and justifies these conclusions. It does not address the question of the worker's acceptance of these devices because at the present time, no device is sufficiently advanced for this question to be meaningful.

RÉSUMÉ

À partir de l'examen approfondi de la documentation récente traitant des dosimètres passifs de produits de filiation du radon, nous avons conclu que :

(1) des dosimètres passifs pour mesurer le radon sont disponibles et sûrs;

(2) à l'heure actuelle, il n'existe pas de dosimètres passifs acceptables pour mesurer les produits de filiation du radon. Dans un avenir prévisible, il existe bien peu, sinon aucun espoir de voir la mise au point de tels dispositifs;

(3) les capacités des «dosimètres semi-passifs» sont plus que douteuses, mais nous sommes moins résolus à déclarer catégoriquement que ces dispositifs ne peuvent être mis au point pour mesurer les produits de filiation du radon de façon utile.

Le présent rapport documente et justifie ces conclusions. Il ne traite pas de la réaction favorable ou non des travailleurs à ces dispositifs car, en ce moment, aucun dispositif n'est suffisamment à point pour que la question vaille la peine d'être abordée.

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1. INTRODUCTION

It has been recognized for some time that exposure to short-lived radon (Rn-222) progeny (commonly called radon daughters) is the major cause for the increase in the incidence of lung cancer in some groups of uranium and other miners. This recognition has led to the reduction of exposure limits for radon daughters, from essentially uncontrolled levels at the start of the uranium mining boom in the early fifties, to current limits ranging from about 3 to 5 working level months (WLM), depending upon jurisdiction. The recognition of the hazard of radon daughters has also resulted in a need for better radon daughter concentration (WL) and radon daughter exposure (WLM) measurements.

Traditionally, WL has been measured using "grab sample" techniques, where a sample of air was drawn through a filter and the measured alpha activity on the filter was used to estimate WL by standard (usually Kusnetz) methods. A miner's WLM could then be estimated from these measurements by multiplying the average WL measured at the different work stations by estimates of the number of hours a miner worked at that station, and then summing over the results for all the work stations. Since it is known that the WL can vary considerably at any given location, and is very dependent on local conditions in a mine -- that is, there is a large temporal and spatial variability — there has been some expressed concern that grab sampling does not give results representative of an individual miner's exposure. This concern has led to the development of other methods of estimating exposure, most notably continuous area and personal radon daughter exposure monitors.

There has been considerable work on the development and testing of personal radon daughter exposure monitors (usually called dosimeters) since the mid 1970's. These are viewed as the most promising way of obtaining representative exposure measurements for miners. These personal dosimeters are often grouped into active (those with a mechanical pump) and passive (those without a mechanical pump). Active dosimeters have received the most attention, mainly because of the known or perceived difficulties of collecting airborne ions and aerosols without a mechanical pumping system. Various active personal radon daughter dosimeters have been developed and tested in Canada and other countries. While some of these dosimeters have shown considerable promise, they are not in widespread use. The major criticisms have been poor reliability (usually associated with the mechanical pump), weight and size of the detector pump and battery system; and cost.

In principle, passive radon daughter dosimeters have the potential to overcome most of the criticisms of the active dosimeters and much attention has been devoted to them in recent years.

Passive dosimeters suffer from known and perceived problems, notably poor sensitivity, large variability in results for constant radon daughter concentrations, and an inability to withstand the harsh environments encountered in a mine. Various methods have been tried to overcome these

problems. Examples of attempts to increase the sensitivity include the use of high sensitivity (track etch) detectors and electrostatic collection. The method most often used to remove variability in results is to use a diffusion barrier. However, this barrier results in a dosimeter that only responds to radon and the degree of disequilibrium which is necessary to determine WL must be obtained by some other means.

1.1 Types of Personal Radon Daughter Dosimeters

To systematically discuss personal passive radon daughter dosimeters, it is useful to classify the various types of personal dosimeters.

Dosimeters can be usefully classified according to the means of sample collection, according to whether they are sensitive to radon daughters or only to radon, and according to what detection method they use. Their sensitivity to thoron and thoron daughters is an important characteristic but we will not use it as a classification.

Sample collection: In "active" dosimeters, sample collection relies on a pump; in "passive" dosimeters, no pump is used. Passive dosimeters can be further divided into purely passive dosimeters and dosimeters which rely on some driving force to augment their natural collection. We will refer to these latter dosimeters as semi-passive.

Radon daughter sensitivity: Ideally, the dosimeter should be sensitive to radon daughters, not just to radon. However, the development of radon only passive detectors is much more advanced than radon daughter passive detectors. A comparison between the two types of dosimeter is necessary.

Detection method: The various dosimeters can be classified according to the detection method used. The two most common detection methods are thermal luminescent detectors (TLD's) and nuclear track detectors. Less common methods include solid state detectors, thermally stimulated exoelectron detectors (TSE) and photographic film.

The most important distinction is among active, semi-passive and passive dosimeters. It determines whether the dosimeter will be sensitive to radon or not, and whether a TLD is an appropriate detector.

1.1.1 Active Dosimeters

In an active dosimeter, a flow of the radon daughter laden air is established by means of a mechanical pump. This flow passes through a filter which traps the radon daughters regardless of whether they are attached to aerosols or not, but which allows the radon to continue through. The radon daughters decay in place on the filter and these decays are recorded. The total activity of the collected daughters and their progeny

is determined. From a knowledge of the flow rate, the working level (WL) exposure can be derived. Because the daughters are trapped and the radon is not, an active dosimeter's sensitivity to radon is very small compared with its sensitivity to daughters.

Active dosimeters can use either a TLD or a track detector for measurement. Other detectors are also sometimes used.

When a nuclear track detector is used, the energy of the emitted alpha particle can be measured by the use of absorbers or by measuring the etch pit parameters. Its origin from either RaA or RaC' can then be established, which allows the Working Level to be determined. Even if the energy of the individual tracks is not determined, a reasonable estimate of the working level can still be obtained. Note that the number of tracks recorded from a particular isotope is not proportional to the activity concentration of that isotope in the sampled air. Rather it is proportional to the number of decays of that particular isotope regardless of the identity of the parent isotope when trapped. Because the collection time is long compared to the lifetimes of the short-lived daughters, the number of counts is directly related to the total alpha energy deposited. This is distinctly different from nonintegrating measurement techniques, where the total potential alpha energy is calculated from the individual concentrations of the various daughters.

Working level is defined in terms of the total potential alpha energy in a volume of air. Active radon daughter dosimeters measure this energy directly.

Since a TLD responds directly to the total energy deposited, its response will be essentially proportional to the radon daughter working level with only minor, if any, corrections required. However, if significant and variable quantities of thoron and thoron daughters are present, the utility of TLD's is somewhat compromised. This is because, when assessing radiological hazards, a working level of thoron daughters is less heavily weighted than a working level of radon daughters. There is no simple way to differentiate between these two with TLD's.

The perceived problems with active dosimeters include cost, reliability, and the uncertainty in the effective flow rate. However, the performance of active dosimeters has been considerably improved in the last few years and may now be entirely satisfactory.

1.1.2 Passive Dosimeters

By definition, passive dosimeters lack a pump. Lacking a pump, they do not collect radon daughters on a filter for measurement. Instead they measure the radon daughter concentration in some defined volume of air. Depending

on the design of the dosimeter, this volume might be defined by the range of the alpha particles in air or it might be defined by the enclosure. This volume source method has a number of immediate consequences -- most of them undesirable.

Because the geometry of the emitter relative to the detector is not fixed, the detected energy does not define the identity of the emitter. It might be a low energy alpha originating close to the detector, or a high energy alpha from a greater distance. In principle, if the energy of each detected alpha were measured, and if the expected spatial variation of the concentration of the various daughters over the detection volume were known, then it might be possible to unfold the measured spectrum and determine the concentrations of the individual daughters present. Since TLD's record only the total energy deposited, TLD's do not provide sufficient information to allow the working level to be determined, even in principle.

Passive radon daughter dosimeters do not measure the working level directly. The working level must be derived from the concentrations of the individual daughters, but only the alpha emitting isotopes are detected. Thus a passive dosimeter cannot give any information on the concentration of the beta emitter RaB which contributes significantly to the working level. This missing information affects the accuracy of the final answer and is discussed more fully in Appendix A.

Passive radon daughter dosimeters will also be sensitive to any daughters which plate out on the detector and perhaps on the walls (if any) of the defined volume. In general, there is no simple relationship between the amount of activity plated out and the activity in the air. It will be dependent upon the nature and concentration of the aerosols, the temperature and the humidity. Also the plating out of daughters directly onto the detecting surface effectively reduces the concentration in the air directly above that surface. The concentration of a particular daughter is not uniform throughout the sampling volume. Much of the work on passive radon daughter dosimeters has focussed on ways to overcome these plate out effects. (The plate out effect is a much less serious problem in active dosimeters because the plate out location is largely determined by the forced air flow.)

Because passive radon daughter dosimeters are measuring the alpha activity in a volume of air, as opposed to on a filter, they will also register alphas from radon. Since the radon activity in air is always greater than either the RaA or the RaC' activity and is never significantly less than the combined activities, this is a significant interference. Many passive radon daughter dosimeters, make a separate determination of the radon concentration in order to correct the working level concentration for the contribution from radon.

The detector in a passive radon daughter dosimeter must be exposed directly to the air. The detector is therefore susceptible to fouling by whatever

happens to be in the air — dust, mud, explosive powders, acid sprays, diesel fumes, oil mists, etc. If the detector is protected, its sensitivity to radon daughters is greatly reduced. If it is soiled by an unknown amount of dust or liquid, its sensitivity is unpredictable.

1.1.3 Semi-Passive Dosimeters

Semi-passive dosimeters are pumpless dosimeters that augment the collection of daughters by some other means — electrostatically for example. In all cases that we are aware of, the daughters are collected onto a surface rather than concentrated in a volume. The detection characteristics of these dosimeters are therefore closer to those of active dosimeters than passive dosimeters.

The major difficulty with semi-passive dosimeters is the large uncertainty associated with the effective pumping rate.

1.2 A Brief Summary of The Physics and Dosimetry of Radon and Thoron Daughters

Radon and thoron daughters are recognized as presenting a significant radiological hazard in underground mines -- especially in Uranium mines -- and in other locations. Radon (Rn-222) is a consequence of the presence of U-238 in the rock, whereas thoron (Rn-220) is a consequence of the presence of Th-232 in the rock. Indeed, the presence of radon is sometimes used in seeking uranium deposits. A simplified decay series for these two species is shown in figures 1 and 2. Because radon is a gas at normal temperatures, it can escape from the rocks and pervade the underground atmosphere. When radon or thoron decays in the air, it leaves a (usually) charged airborne daughter. These daughters are not gases at environmental temperatures and will tend to plate out on any solid particle, surface, or aerosol that they encounter. Because these daughters are also radioactive, they too will decay until eventually a stable daughter is reached.

It is generally accepted that the airborne radiation exposure of the human lung is predominantly due to the short-lived radon (and thoron) decay products filtered out of the inhaled air in the respiratory tract and not to the radon (thoron) itself. Accordingly, the working level is defined in terms of the potential alpha energy that will be delivered to the lungs due to the daughters which are deposited in the lungs. Radon and thoron do not contribute to the working level because they are not filtered out in the respiratory tract.

One working level (WL) is defined as any combination of the short-lived decay products of radon (Po-218, Pb-214, Bi-214, and Po-214 — often referred to as RaA, RaB, RaC, and RaC') in one litre of air that will result in the ultimate emission by them of 1.3E5 MeV of alpha energy. A working level for thoron is similarly defined. However, the exposure limits, in terms of working level months, is about three times higher for thoron than it is for radon. This results from the fact that, due to the long half life of ThB (Pb-212), some of the thoron daughter products are cleared from the lungs before they can decay.

The concentration of the various daughters in the air, for a given radon or thoron concentration, is a function of the rate at which the daughters are being removed from the air via plate out and of the "age" of the air.

Ignoring plate out, the build up of the daughters as the air ages is presented in figure 3 for both radon and thoron. Figures 4 and 5 show the build up of their contribution to the working level.

A detailed description of the build up of radon daughters is presented by Evans [1]. Much of that discussion can be directly applied to thoron by simply changing the numerical values of the decay constants and alpha energy.

2. COLLECTION METHODS

The means of sample collection is of fundamental importance in characterizing passive radon daughter dosimeters. Purely passive dosimeters rely on diffusion and/or conventional turbulence to ensure that the volume of air being measured is representative of the ambient air. Semi-passive dosimeters rely on some physical force to augment the collection. In this case, the aerosols are usually collected on a surface to be counted. The driving force behind the collection is usually the consequence of maintaining a gradient in some physical property. For example, diffusion results from a concentration gradient. Electrically charged particles and molecules can be collected by an electric field (the gradient of electric potential). Polarizable uncharged aerosols can be collected with an electric field gradient. Thermophoresis is the collection of particles due to maintaining a thermal gradient. In principle magnetically polarizable particles could be collected using a magnetic field gradient. It is likely that there are other examples — humidity gradients, perhaps.

We will explicitly discuss diffusion, electric fields and thermophoresis.

2.1 Diffusion

Passive monitors are now in extensive use for measuring the exposure to many organic vapours. These monitors rely on controlled diffusion into the detection volume. The design is such as to attempt to provide independence from external wind velocity effects. The mass flux of vapour into the detector can be calculated from Fick's First Law. That is, the mass flux of vapour is proportional to the area of the inlet opening, the concentration difference between inside and out and inversely proportional to the distance the vapour must diffuse before detection. The constant of proportionality is the diffusion coefficient. For a recent discussion see Posner and Moore [2].

Diffusion can lead to controlled collection for molecular and atomic species where the diffusion coefficient is large and well defined. For aerosols, the diffusion coefficient is much smaller, is size dependent, and in general, will not be known.

For a passive dosimeter, a knowledge of the effective diffusion rate is not necessary. This is because the measurement is not flow rate dependent. All that is necessary is that the radon daughter concentration in the volume being measured is in equilibrium with the ambient concentration. For an exposed detector this does not impose any restrictions. If the detector is protected from dirt in the environment, the diffusion rate might be limiting in the amount of protection that could be provided.

2.2 Electrostatic

Electric fields are often used in passive radon dosimeters to collect the decay products after the radon has diffused through a permeable barrier and then decayed within a collection enclosure. In this case, the electric field is collecting unattached charged radon daughters in an essentially aerosol free environment. In addition, some passive radon dosimeters use a desiccant to keep the relative humidity in the collective volume low.

The use of electrostatic collection for ambient radon daughters is less common. In this case, there is no effective control of the environment.

Kahn and Phillips have utilized electrostatic collection for the direct collection of ambient radon daughters [3-4]. In that work, the electric fields originated on an electret. In a subsequent paper [5], they investigate the effect of environmental conditions on collection efficiency. For experimental ease, charged plates were used instead of electrets.

In reference 3, they reported that the rate at which radon daughters are collected increases linearly with voltage over the range studied and, for their experimental conditions, that a potential of 240 volts is all that was necessary to equal the pumping speed of a CEA active dosimeter. However, they also found that the collection efficiency is strongly dependent on humidity. At a given humidity, the collection rate was reproducible to within 10%.

They also found that uncharged aerosols could be collected. The collection of uncharged aerosols is not unexpected as it is due to the induced polarization interacting with the electric field gradient.

Reference 6 extends the results of reference 3 to an actual practical dosimeter. This dosimeter is further described in 7. The conclusion is that the results for these semi-passive dosimeters are encouraging, but more work is necessary.

The major problems appear to be the charged fraction dependence and the humidity dependence of the collection efficiency. Their results show that there is a variation of greater than a factor of two as the relative humidity ranges from 40 to 100%. However, the significance of this variation can only be judged on the basis of the humidity variations encountered in real mines. It is also perhaps significant to note that the collection efficiency is most sensitive to humidity at higher relative humidities.

The other potential problem involves the different collection efficiencies for charged and uncharged aerosols. The observation [5] that the various daughters are pumped at different rates suggests that the charged fraction is definitely affecting the collection rates. This result is not unexpected and this is another parameter that must be known, or its effects shown to be small under typical mine conditions. Reference 5 presents some data indicating the latter.

The experimental data discussed above were collected on a disc directly exposed to the air of an experimental radon chamber. A practical dosimeter would require some protection from the hostile mine environment. It seems likely that this would reduce the collection efficiency somewhat.

In reference 4, Khan and Phillips investigated the efficacy of electrostatic collection based on the use of an electret in a configuration more representative of a practical dosimeter. In this case, the collection efficiency was notably reduced; potentials of the order of 2000 v were necessary to equal the performance of the CEA dosimeter.

The present authors have difficulty in imagining how to protect the collection surface from the outside environment without enclosing the dosimeter. Enclosing the dosimeter with an insulating surface will provide protection and will allow the electric field lines to extend outside the enclosing surface, but it is unlikely to provide a reproducible collection efficiency. Charges in the environment will collect on the insulating surface and will tend to neutralize the external collecting field. If protection is provided by a conducting surface, there will be no external fields. In this case, the dosimeter will be relying on diffusion of the daughters to the entrance holes in the shield.

Cowper and Davenport [8] have reported the humidity dependence of their passive radon dosimeter. They also summarized the results reported in the literature to that time and note that there are large discrepencies in the reported effects of humidity.

A recent paper by Bigu [9] discusses the effect of electric fields on Rn-220 (thoron) progeny but his primary interest was concerned with controlling the atmospheric thoron daughter concentration with electric fields. However, some of his observations are relevant to the present discussion. In

particular, he notes that "the decrease in thoron progeny concentration by the application of a negative voltage of the test facility electrodes is consistent with two well documented facts, namely: (a) a large fraction of thoron progeny aerosols are electrically charged; and (b) thoron progeny are initially formed in a free, positively charged state. From the dramatic decrease in thoron progeny concentration in the presence of the electric field, it may be concluded that a large fraction (60%) of the radioactive aerosols are electrically charged. However, the observation that even much higher potentials do not reduce the thoron progeny concentration to a negligible amount indicates that a significant fraction of the thoron progeny are in a neutral electrical state."

Kotrappa et al. [10] discuss the use of electrets for the passive detection of radon. The electret is teflon with a surface charge of 1-3 kV. The methods relies on the decay products having a positive charge. In another paper by the same authors [11], they describe similar work. They find that for radon daughters the collection efficiency decreases with rising humidity, but for thoron daughters, the variation is the other way. They cite other work which found the same thing.

These authors also describe an active dosimeter for radon daughters based on an electret [12]. In this dosimeter a pump draws a measured sample through a filter with an electret mounted above the filter on the inlet side. The loss of charge on the electret is a measure of the alpha decay within the volume. Because that volume is small compared with the total volume which passes through the dosimeter, most of that alpha activity is from the filter. The paper does not address the performance of this dosimeter in adverse conditions.

2.2.1 Electrets

The above discussion has centred largely on the utility of using an electric field to enhance the collection of radon daughters. It was largely immaterial as to how that electric field was produced; the response of the aerosols should be the same. However, for a practical dosimeter, there is a great advantage to being able to produce the field without the use of external sources of power. Electrets appear to be a practical means of doing this.

A comprehensive modern review of electrets in general is given by Sessler [13]. The following is extracted from the introduction:

"An electret is a piece of dielectric material exhibiting a quasi-permanent electrical charge. The term 'quasi-permanent' means the the time constants characteristic for the decay of the charge are much longer than the time periods over which studies are performed with the electret.

"The electret charge may consist of 'real' charges, such as surface-charge layers or space charges; it may be a 'true' polarization; or it may be a combination of these. ... While the true polarization is usually a frozen-in alignment of dipoles, the real charges comprise layers of trapped positive and negative carriers, often positioned at or near the two surfaces of the dielectric respectively. The electret charges may also consist of carriers displaced within molecular or domain structures throughout the solid, resembling a true dipole polarization. ... Mostly, the net charge on an electret is zero or close to zero and its fields are due to charge separation and not caused by a net charge.

"... an electret is thus in a sense the electrostatic analogue of a permanent magnet, although electret properties may be caused by dipolar and monopolar charges while magnetic properties are only due to magnetic dipoles. The existence of an external field and the corresponding analogy with a magnet has often been used to define the electret.

It may be useful to note that electrets can be used in two distinctly different ways in dosimeters. Firstly, the external fields from an electret may be used to enhance the collection of radon and radon daughters. Devices utililizing this effect are described in references 3, 6, 10, and 11. The first two references refer to a passive radon daughter dosimeter while the last one is specifically for radon.

Secondly, electrets can be used as the actual detector. In this case the electret is supplying a polarizing field to form an ion chamber. The ionization current neutralizes the electret charge. The loss of polarization is then a measure of the time integrated ionization current — that is, the total charge released in the collection volume. Reference 12 is an example of this use in an active radon daughter dosimeter.

2.3 Thermal

2.3.1 Thermophoretic

In the presence of a temperature gradient, an aerosol will tend to drift down the gradient and deposit on the colder surface. This thermophoretic collection of radon daughters has been discussed by Leung and Phillips [14]. While these authors have demonstrated that the method works, they also admit that the method does not seem promising for use in a practical personal dosimeter. The problem is largely one of maintaining a suitably large temperature gradient. Even if this problem were to be solved there still remains the question as to how well it would work under varying environmental conditions.

3. DETECTION METHODS

As previously noted, the choice of detector is dependent on the method of collection. That is, TLDs are usable in active and semi-passive dosimeters, but are less suitable in purely passive devices. On the other hand, track detectors can be used in either type of dosimeter.

3.1 Track Detectors

A nuclear track detector is a material that will be damaged by energetic particles in such a way that the track of the particle can be made visible by appropriate etching techniques.

For a track to be recorded, the particle must produce sufficient damage along the track; it must be of high LET. Gamma rays and electrons are not seen. Less obvious is that there is also a high energy cut off for alpha particles. However, as the high energy particle slows down while penetrating the track detector, it will begin to leave a track. It follows then, that different energy tracks can be brought out by different etching conditions. The higher energy tracks can be developed and counted if sufficient surface material is etched away. However, some of the low energy tracks may be completely lost before the high energy tracks become visible.

Many different etching recipes have been published. Most involve heated, highly basic solutions and etching times of many hours. Sometimes the chemical etching is augmented by the application of an electric field i.e. electrochemical etching.

For an overall review of solid state nuclear track detectors, see Hepburn and Windle [15].

Passive dosimeters for radon or for radon daughters based on the use of nuclear track detectors are of two basic types, those sensitive to radon only, and those sensitive to both radon and radon daughters. The simplest dosimeter responds to both radon and radon daughters. In one manifestation, the dosimeter consists of a track detector exposed to the ambient air. As such, the track detector will record tracks from radon and thoron, from airborne radon and thoron daughters, and from those daughters plated out on its, or adjacent, surfaces. It will also respond to ore dust. The active surface of the dosimeter is subject to fouling.

In the radon only dosimeter, the track detector is completely enclosed. The contact with the environment is through a diffusion barrier. Radon and thoron as gases can diffuse through the barrier, but the daughter products and other interferences are excluded. The dosimeter therefore responds only to the radon (or thoron) content of the air. The track detector will, of course, record the alphas from the daughter products that originated from radon and thoron decay within the diffusion chamber.

Because the counting environment of a radon only dosimeter is better defined and because dirt and other interferences are excluded, these dosimeters perform better than those sensitive to the ambient daughters.

By the appropriate choice of diffusion barrier, the diffusion rate can be slowed down to the extent that all of the thoron will have decayed away before reaching the interior of the dosimeter. By varying the choice of diffusion barrier, different degrees of sensitivity to radon verses thoron can be achieved.

The literature concerning passive radon dosimeters is much more extensive, presumably because the problem is much more tractable. Some representative articles are references [10] and [16-36].

The earlier nuclear track detectors used in radon and radon daughter dosimeters were based on cellulose nitrate. Depending on the actual plastic and the etching conditions, the maximum energy at which an alpha particle would produce an etchable track is in the range 2 to 5 Mev. This is below the alpha energy of RaA and RaC'. This has the desirable property that this detector is insensitive to radon daughters plated out on its surface. The detector is also insensitive to full energy radon alphas, but not to degraded radon alphas.

CR-39, however, is a more sensitive track detector. It responds to alpha particles up to almost 8 Mev. CR-39 will therefore respond to plated out daughters. Because the plating out of daughters is, for all practical purposes, essentially an unpredictable process, it has become conventional wisdom that CR-39 is unusable in a passive dosimeter where it is directly exposed to the environment. However, it is now commonly used in radon dosimeters; that is, in situations where it is protected from the environment by a permeation barrier. In this case, the plate out is more predictable and the higher sensitivity can be put to good use.

The present authors feel that the non-utility of CR-39 as the naked detector should be re-examined. For example, some recent results by Cross et al. [37] show that the high energy cutoff of CR-39 can be readily controlled by the appropriate choice of pre-etching and subsequent electrochemical etching. Energy analysis based on etch pit size could also be used [38-41]. This maybe somewhat academic, however, since we also feel that there is little real hope for a passive dosimeter that employs an uncovered nuclear track detector exposed to the mine environment.

A fairly thorough early review of some passive plastic nuclear track detector (NTD) systems for radon and radon daughters is given by Frank and Benton [21]. They describe the general nature of the radon problem and discuss various options for exposure measurements. They concluded that the active dosimeters then available could not perform satisfactorily in a mine environment.

Passive NTD dosimeters do not rely on collecting the daughters. Instead they record the alpha particles emitted by airborne radionuclides. They, therefore, have the disadvantage of being sensitive to Rn-222 and have less absolute sensitivity. For passive dosimetry, nuclear track detectors have more favourable characteristics than TLDs. TLDs are rather insensitive to ambient alpha particles in the presence of beta and gamma backgrounds. When exposed to ambient air, as they must be, they are sensitive to plated out material on or near their surfaces. Some nuclear track detectors are insensitive to alphas of these energies. However, alphas from uranium ore dust can cause tracks which are registered.

Track etch detectors can be used in both active and passive devices but they compare more favourably with other detector methods in passive devices. For active sampling, these authors prefer TLD's. Track etch detectors are of adequate sensitivity but readout is less convenient, requiring wet lab etching and track counting.

The bulk of the paper [21] then goes on to discuss a particular dosimeter, designated the USF passive track-etch dosimeter, in detail.

Nuclear track detectors are discussed in a subsequent paper by the same authors [42]. By this time they had abandonned cellulose nitrate in favour of CR-39 where possible. Their introduction is a good summary of the passive nuclear track detector system. We will quote the first two paragraphs in their entirety.

"The selective sensitivity of plastic nuclear track detectors (PNTD's) to low energy alpha particles in an environment which also contains gamma and beta radiations has made these detectors prime candidates for the dosimetric measurement of the concentrations of radon and its daughter products in mine air. Their passive, integrating mode of measurement, small size, low weight and inexpensiveness are attrative characteristics for large scale personal dosimetry.

"The detectors can be used in either active or passive dosimeters. In active devices, the PNTD is placed in close proximity to a sampling filter. The filter collects, from a calibrated air flow, all the daughter nuclei which are in suspension. As the daughter nuclei pass through the decay chain through Po-214, a fraction of the alpha particles emitted are registered as latent tracks in the plastic. In passive devices, the PNTD is placed in direct contact with the ambient air containing the radionuclides concentrations to be determined. The active dosimeters have the advantage of excellent sensitivity, and the measured track densities yield very close approximations to accumulated Working Level (WL) exposures. They have the disadvantage that the simplicity of the passive PNTD is lost, since a battery-driven constant flow-rate air pump is a necessity. The passive dosimeters are simple in construction and use, but they have the

disadvantage that WL exposures are not directly measured and certain assumptions concerning radon and daughter equilibrium conditions must be made in determining WL exposures from the measured track densities. Also the sensitivities, in track densities per Working Level Hours (WLH) exposure are much less than for active dosimeters."

Because PNTD's are sensitive to Rn directly, measurements using PNTD's directly exposed to the ambient air are very sensitive to the ratio of radon to radon daughter concentrations. Furthermore, the detectors do not weight the individual daughter activities in proportion to their importance to WL. Reference 42 addresses the problem by using a dosimeter which measures the Rn present separately. Electric fields were used to enhance the collection inside the diffusion chamber used to measure radon. The disadvantage of this method is that it has a humidity dependence.

For measurement of the ambient alphas (i.e. no diffusion barrier) the most suitable PNTD found is cellulose nitrate plastic. CR-39 is not used because it is sensitive to alpha particles emitted by daughter nuclei which plate out on its surface. They found that a simple exposed piece of plastic did not work because of the dirt and dust build up. They tried shielding the detector by recessing it. The result was that the response to the daughters decreased by much more than the response to radon. They conclude "that there is no acceptable trade off between the shielding effect and the contamination problem, since relatively small amounts of shielding degrade the detector response while a more substantial amount of shielding would be necessary to adequately protect the PNTD from contamination. The possibilities of different shielding configurations have been considered, but none have yet been devised which would not discriminate against ambient daughter activities."

To determine WL exposure from paired ambient and radon detectors, it is necessary to relate the ratio of the measurements to a standard model of radionuclide equilibrium conditions. No information is collected about the relative abundance of the various daughters.

Domanski and his colleagues have been very active in the development of passive radon/radon daughter dosimeters for use in Polish mines [19,20,43-49]. These dosimeters have used nuclear track detectors.

The various dosimeters and their use in Polish mines is presented in references [19, 47, and 49]. Basically, the dosimeter consists of a piece of nuclear track detector, originally cellulose triacetate film and later LR-115. This detector is fitted into a small cassette which affixes to the back of the miner's helmet. It is open to the environment so that it records the radon and radon daughter alpha particles emitted in the air. Because of the choice of track detector, it

does not respond to daughters plated out directly on it. The calibration, that is, the number of tracks as a function of working level, is based on the assumption of a particular ratio between radon level and working level. This is the primary source of the ± 30% uncertainty in these measurements [46]. These papers do not address the question of the response of these dosimeters to the dirt and grime of the underground environment.

The other papers by Domanski et al. are concerned with the theory of using nuclear track detectors for radon and radon daughters. References 44 and 45 describe their original measurements on LR-115 nitrocellulose foil. These results show the utility of LR-115 but also show its sensitivity to the radon/radon daughter equilibrium ratio.

In reference 46, Domanski et al. hoped to improve the performance of a single foil dosimeter by the use of multiple absorbers for energy discrimination. In principle, this should allow the equilibrium ratio to be determined. Unfortunately, the experimental data did not support this hope.

In reference 19, Domanski et al. discuss the usefulness of providing a track detector protected by a diffusion barrier so as to measure the radon concentration. This datum can then be used to correct the ambient detector measurement. They conclude that such a differentiating track detector is not very sensitive to the relative quantities of radon daughters and radon in the surrounding air. In practice, it cannot provide sufficiently precise information about the equilibrium ratio to be useful. The problem is particularly acute for very young air. This theme is also presented in reference 48.

In reference 20 Domanski et al. present a detailed discussion of plate out and how it affects the choice of track detector. They conclude that the critical energy of the ambient detector should be less than 6 Mev. This would appear to eliminate CR-39. Cr-39 would also be more sensitive to radioactive ore dust. They conclude that it is questionable whether CR-39 is suitable for use as a single detector passive dosimeter for radon daughters. They believe that it can be successfully used in diffusion detectors as the inner bare film in the diffusion chamber. In that use, its high critical energy will enhance the detector's sensitivity without its utility being destroyed by plate out.

Pfligersdorffer et al. [50] discuss both CR-39 and LR-155 with respect to the measurement of environmental levels of radon and its decay products. They conclude "Nuclear track detectors can be applied successfully to the measurement of low level Rn concentrations with a lower detection limit of about 0.1 pCi/l for an integration period of three months. There are no great differences between LR-115 and CR-39." While the experimental set up included an exposed detector in addition to one protected by a diffusion barrier, the authors did not address the problem of daughter determination except to point out that exposed LR-115 registered fewer tracks than exposed CR-39. This was attributed to the high energy cut off of LR-115; that is,

plated out daughters did not register on the LR-115 because their energy was too high. Only airborne daughters and Rn-222 registered. The authors did not comment on the usefulness of this response in a practical dosimeter.

The Terradex Corporation has been active in developing radon and radon daughter dosimeters based upon the use of nuclear track detectors. Some representative references are: 16, 17, and 51-54. In reference 17, Alter et al. describe some applications of a Terradex sampler. They also explain some considerations relevant to the choice of membrane. They did not explicitly identify the Terradex membrane but stated that it delays the entry of radon isotopes by 2.5 days with the result that the short-lived radon isotopes Rn-220 and Rn-219 decay before they cross the membrane. Metal atoms and aerosol particles are totally excluded. Only Rn-222, because of its longer mean life (5.5 days) is able to penetrate significantly. This membrane allows the interior level of radon to reach 65% of the outside value. It has the additional virtue that water vapor penetrates only slowly. Highly permeable experimental membranes can also be used to enhance [relative to less permeable membranes] the signal when low levels of radon are present. In general these membranes will also show an increase in the permeation of water vapor which makes them unsuitable for some applications.

Another covering option is a simple filter. This allows gases to enter the detection space freely, but exclude aerosols and metallic radionuclides. In this case both radon and thoron are recorded if present, and their concentrations inside the enclosure are identical to those outside. The presence of thoron can be recognized by simultaneous measurements using a filter covering and a membrane covering.

In reference 54, Gingrich et al. propose passive radon monitors for radon daughter measurements on the hypothesis that the ratio between radon and radon daughters is consistent enough on an average basis to convert from radon measurements to WL measurements with a smaller uncertainty than that obtained via grab sampling.

In reference 52, Fleischer et al. describe some passive measurements of working levels by the nuclear track technique. The method works by a passive measurement of the radon concentration and a passive measurement of the working level ratio (WLR). The paper then goes on to describe some of the problems of plate out. If radon daughters were uniformly distributed in space, the problem of making passive long-term WLR measurements would be simple. Because the daughters plate out on surfaces, the concentration on the surface is high, but the concentration in air near the surface is lowered. Since any measurement method involves surfaces, i.e. the detector, the very act of measuring disturbs that which is being measured. The depleted concentration near the surface can only be calculated if the effective diffusion constant is known. But this is environment dependent. These plateout considerations are what makes the two-parameter methods for determining radon-daughter concentrations fundamentally invalid. The method presented in [7] involves the five

separate parameters. Rn-222 is measured by the standard nuclear track detector/diffusion barrier method. Four other nuclear track detectors are provided. Three of these are covered with different thicknesses of polycarbonate absorbers. Good correlation is obtained between the usual active WLR measurements and the proposed passive ones as long as a reasonably well defined one dimensional diffusional environment is maintained. These results have been obtained in clean conditions. Its use in dusty mine conditions will have to be evaluated. The device as described is to be used as an area monitor. The underlying theory is presented in reference 51.

A recent study of plate out is presented by Wong [55].

Kahn et al. have written several papers on the general subject of nuclear track detectors for radon and thoron [56-59]. In reference 56 they studied the characteristic differences between the etch pits due to radon and thoron alpha particles in CA80-15 and LR-115 cellulose nitrate track detectors. They concluded that "both the development and the annealing properties of latent damage trails due to the alpha particles produced in the decay of radon are different from those produced in the decay of thoron" and that "CA80-15 and LR-115 cellulose nitrate detectors can be effectively employed in discriminating between radon and thoron in their mixed atmospheres. This can be used in separate estimations of radon and thoron contributions in mines." Reference 59 presents similar results as well as discussions of the utility of using these detectors for "dosimetry, prospection [sic], and for the discrimination between uranium and thorium ore bodies." These papers do not add any new dimensions to the present discussion. Reference 58 covers the same ground as reference 59.

Reference 57 presents the results of their preliminary investigation of CR-39. They conclude that CR-39 detector is much more sensitive than the previously used detectors for radon/thoron personnel [personal] dosimetry and that the damage tracks are highly stable with respect to temperature and humidity effects. "CR-39 appears to be the best available detector for radon/thoron personnel [personal] dosimetry."

Many of the more sophisticated approaches to measurement using NTDs require energy discrimination. In a recent article Fews and Henshaw [39] describe a method of "high resolution alpha particle spectroscopy using CR-39 plastic track detector." The method consists of inferring the alpha particle energy by means of measuring a number of parameters characterizing the etch pits. A somewhat less sophisticated analysis method has been investigated by Pai [38]. Wong and Tommasino [40] and Espinosa et al. [41] report other methods of achieving the same thing. In principle, all of these methods could be extended to automatic processing, but with difficulty. However, the discussion of Appendix A suggests that even energy discrimination may not be sufficient to produce a dosimeter with the appropriate response.

3.2 Thermoluminescent Detectors

Nuclear track detectors are more appropriate than TLD's for passive radon daughter dosimeters. Most of the modern work concentrates on track detectors. However, there has been some discussion on TLD's.

A description of two passive radon and radon daughter dosimeters based on the use of TLDs is given by Niewiadomski and Ryba [60]. While they produced a working passive monitor, its sensitivity was very low. They claim that the advantage of TLD based systems is that the TLDs respond to dose (as opposed to number of events) and therefore they they are insensitive to the equilibrium factor (but they remain sensitive to the radon directly). They support this claim by presenting experimental data based on collecting radon daughters onto a filter. There is no mention of what the implications are when the source is a volume of air.

A disadvantage of TLDs is their sensitivity to gamma rays and beta particles. Practical dosimeters therefore require a background measuring chip as well.

McCurdy et al. [61] discuss the relative merits of three TLD materials, LiF, CaF_2:Dy, and $CaSO_4$:Mn, in active dosimeters. Phillips also addresses this question [62].

3.3 Solid State Detectors

Junction detectors can be used in passive dosimeters to provide energy discrimination. While such a dosimeter is of necessity much more complicated it is one of the few feasible methods of providing the energy discrimination necessary to unambiguously interpret the dosimeter response. Such a dosimeter is much more convenient with respect to read out and can be read daily. Such a dosimeter is described by Bigu [63].

3.4 Thermally Stimulated Exoelectrons

Dosimeters based on thermally stimulated exoelectrons (TSEE) have been studied for use in radon monitoring by Gammage et al. [24,64]. We are not aware of any instance where they have been used for radon daughters.

Since TSEE responds to dose, they are more likely to be of use in an active or semi-passive radon daughter dosimeter than in a passive one.

According to private conversations with A.R. Jones, there has been significant improvement lately in the field of exoelectron emission. For a key to the recent literature, see the recent special edition of Radiation Protection Dosimetry [65].

3.5 Photographic Film

Radon and radon daughters can also be detected by photographic film. Bedrosian describes a device [66] that utilizes ZnS to convert the alphas to light pulses which are recorded on Polaroid film. Two detectors, one protected by a filter and one exposed to the atmosphere are provided so as to differentiate between radon and radon daughters. We know of no recent dosimeters that are based on photographic film.

4. CONCLUSIONS

On the basis of our review of the recent literature we have concluded that:

1) Passive dosimeters for radon are available and reliable, at least for non-mine environments. The problem of protecting a radon only dosimeter is much easier than protecting a radon daughter dosimeter.

2) There does not presently exist an acceptable passive dosimeter for radon daughters. There is little if any hope for the development of such a device in the foreseeable future. The major problem is protecting the dosimeter from the mine environment while still maintaining satisfactory sensitivity and achieving a response which is proportional to working level regardless of the degree of disequilibria. Requiring satisfactory performance in the presence of thoron and thoron daughter products is an additional severe complication.

3) We are pessimistic about the potential of "semi-passive dosimeters" but are less firm about stating categorically that no hope exists. The major hope would appear to be the use of an electric driving force. It seems however that protecting the detector from the mine environment while still maintaining sufficiently predictable collection regardless of humidity and aerosol size, density, and charge state will be very difficult, if not impossible.

5. DISCUSSION

The above conclusions are consistent with previous assessments.

In late 1979, H. Stocker prepared a report entitled Personal Radon Daughter Dosimetry — The State of the Art [67]. With regard to passive dosimeters, this report concluded " Owing to the highly unfavourable conditions of wetness, dustiness, mudiness and of other adverse conditions in operating mine environments, it will be a major challenge to design a passive device that will remain clean yet will have adequate sensitivity for reliable exposure estimation at concentrations of radon (and thoron) daughters usually encountered. To our knowledge, no passive radon daughter dosimeter has been fully developed and shown to be reliable in an operating mining environment." He also presented very similar material in reference 68.

In 1982, Frank and Benton, submitted a report to the U.S. Bureau of Mines entitled "Working Level Dosimetry Using Plastic Nuclear Track Detectors" [69]. They reported "A contamination of the ambient track detector surfaces by alpha-active ore particles resulted in high densities of spurious tracks to be measured. A study of protective shielding of the ambient detectors against contamination showed that shielding is incompatible with accuracy for these detectors. The three-element dosimeters have an improved theoretical accuracy but, in practice, they are subject to the same contamination problems and experimental limitations as the two-element models.

"... radon exposures are measured by track detectors isolated from environmental contamination. The accuracy of these measurements is sufficient for personal dosimetry.

"Active dosimeter measurements demonstrated that a much greater accuracy and sensitivity is possible by this method as compared with passive dosimetry."

A study prepared for the A.E.C.B. by R.A.D. Service and Instruments Limited [70] (H.L. Pai) in 1983, addressed the use of CR-39 nuclear track detector in a passive radon daughter dosimeter. The study considered using electric fields for enhanced collection. The introduction of this report provides a good summary of the status of passive radon daughter dosimeters at that time.

The report begins with a summary of Domanski's work. Pai points out that Domanski followed the 'volume source' and 'total absorption' approach for the design of his passive dosimeter. The volume source method assumes that a monoenergetic alpha source is distributed uniformly throughout a homogeneous medium. If an alpha counter is placed on the surface of a volume source, it detects alpha particles of energy up to the maximum energy of the emitter. Alpha particles are detected in a volume determined by the maximum range of the alpha particle.

A basic requirement of this method is that the source be homogeneous throughout the detection volume. If it is not homogeneous, it is very difficult to estimate the detection efficiency from first principles. But plate out effects give rise to a lower concentration in the vicinity of the surface because the surface is removing daughters from the air. Since plate out is essentially impossible to avoid, it effects must be taken into consideration.

The "total absorption" method is effective in differentiating alpha emitters of different energies, but it also prevents the detection of some energy-degraded alphas from the volume under consideration. This lowers the detection efficiency of the method.

Domanski used LR 115 type II for passive detectors. It has a lower detection efficiency than CR-39 -- by about 1/3. Domanski was pessimistic about improving the accuracy of his methods much beyond 30%. Pai felt that this is due to his use of LR 115.

Pai goes on to summarize the work of Benton. This summary is consistent with the summary presented above.

In summarizing these two works, Pai suggests that the unsatisfactory test results are primarily due to: 1) very low detection efficiency and 2) disadvantages introduced by the volume source method. In view of this, future work should concentrate on 1) improvements in the counting efficiency, 2) simplification of the source geometry, and 3) local calibration with respect to an active dosimeter.

In effect, Pai is concluding that purely passive radon daughter dosimeters are not workable, but there may be hope for a semi-passive dosimeter using nuclear track detectors with enhanced collection with electric fields.

At the Occupational Radiation Safety in Mining meeting held in Toronto in October 1984, C.R. Phillips discussed the metrology of radon and thoron daughters [71]. Part of this talk concerned the then current state of passive dosimetry for radon daughters. He made the following points. Passive dosimetry is now well established for gases and vapours. Sample collection is usually based on diffusion through a stagnant gas layer or permeation through a selective or non-selective membrane. However, only diffusion is applicable for radon daughters. But most radon daughters are attached to aerosols and the diffusivity of an aerosol is very much less than that for a gas molecule. Also, it is size dependent.

Phillips claims [71] that it is now generally accepted that no satisfactory solution exists to the problem of contamination of the detector by dust deposition when the dosimeter depends upon diffusion alone. This is because the collection rate by diffusion alone is so slow.

Electrostatic attraction may be used for aerosol collection. The collection may be due to the induction of a dipole or by virtue of their natural charge. Charged metal discs and electrets have both been used for collection.

Thermophoretic collection is possible but there are practical problems in maintaining suitable temperature gradients.

An important aspect of the design of a collector is to shield it from wind. Otherwise, the amount collected will depend upon the collector's orientation with respect to the wind. Phillips claims [71] that this effect is a significant cause of the scatter reported in previous passive dosimeter tests.

A common theme in most discussions of passive radon daughter dosimeters is that while the performance of these devices is not currently acceptable, the situation for passive radon-only dosimeters is much better. The reproducibility of passive radon daughter dosimeters vis-à-vis passive radon dosimeters is illustrated in the two recent international intercomparisons sponsored by the Commission of the European Communities Radiation Protection Research Programme. These intercomparisons have been reported by Miles et al. [72-74].

They report that for passive integrating radon and radon decay product detectors, "two types of detector were used by laboratories participating in the intercomparison. One type consists of passive track detectors within a container closed by a material that permits Rn-222 to diffuse into the container. These closed detectors provide results that are related to the average Rn-222 concentration existing during the period of exposure. The other type consists of open detectors in which the naked track detecting material is exposed so that it records the alpha particles originating from the decay of Rn- 222 and the decay products Po-218 and Po-214 in room air. The open form of detector gives results that depend on the integrated Rn-222 concentration and on the state of equilibrium between Rn-222 and its decay products within room air."

The results indicated that the closed detectors, that is, radon only detectors, did not reveal any serious systematic errors. However, for the open detectors, there was some data suggesting significant systematic errors, particularly where the equilibrium factor was very low.

For the open dosimeters based on LR-115, the results were more closely proportional to the radon gas exposure than to the radon daughter exposure. For other detectors -- CR-39 and polycarbonate -- which detect plated out daughters as well as radon and radon daughters in the air, the results were proportional to neither radon gas nor radon daughter exposure, but something in between. The results varied strongly with equilibrium factor.

Recently a group of experts of the OECD and NEA [75] reported on the Metrology and Monitoring of Radon, Thoron, and Their Daughter Products. This report, in general, does not editorialize on the utility of passive radon daughter dosimeters, but does, when discussing the work of Frank and Benton note that "in spite of these limitations [plate out effects] though, bare dosimeters may be superior to area monitoring in assessing doses to individual miners working in areas with very different radon concentrations."

In 1980 Stocker stated "... no passive radon daughter dosimeter has been fully developed and shown to be reliable in an operating mine environment [67]. That statement is still true today.

6. ALTERNATIVE METHODS

Since it is concluded that acceptable passive radon daughter dosimeters are not likely to be developed in the near future, it is instructive to reiterate the advantages and disadvantages of alternative methods of estimating exposures to radon and thoron daughters. Selection from these methods should be done with reference to the absolute levels of radon and thoron daughters, their possible spatial and temporal variability, and the cost of the monitoring program. It should be recognized that while there are inherent uncertainties in all of these methods, and that some are inherently better than others, there is also considerable uncertainty in other factors that contribute to the risk from exposure to radon and thoron daughters. It is therefore not very fruitful to demand too high a degree of accuracy in the measurement of WL or WLM if this results in a significant increase in the cost of the monitoring program.

The ICRP has recommended that acceptable accuracy for external monitoring is [76]:

"The uncertainty in the measurement of the annual value of these quantities (or of the upper limits if a cautious interpretation is being conducted) should be reduced as far as reasonably achievable. If these quantities are of the order of the relevant annual limits, the uncertainties should not exceed a factor of 1.5 at the 95% confidence level. Where they amount to less than 10 mSv an uncertainty of a factor of 2 at the 95% confidence level is acceptable. This uncertainty includes errors due to variations in the dosemeter sensitivity with incident energy and direction of incidence, as well as intrinsic errors in the dosemeter and its calibration. It does not include uncertainties in deriving tissue or organ dose equivalents from the dosemeter results."

They do not make specific recommendations for monitoring for internal contamination (of which radon and thoron daughter exposure is a special case), or for air monitoring.

Based on the above, it is our opinion that an adequate monitoring program would be one that would estimate the WLM exposure to the same degree of accuracy as recommended by ICRP for external radiation monitoring.

Possible alternative methods include, area grab sampling, area continuous sampling, active radon daughter dosimeters, semi-passive radon daughter dosimeters and passive radon dosimeters.

6.1 Area Grab Sampling

This method has been traditionally used and, in a properly designed and operated program, can easily achieve the accuracy criteria suggested above. The major disadvantages are that it is difficult to ensure that the sample is representative of the air breathed by miners, as the miner will be

working in places not monitored, and radon daughter concentrations often exhibit large fluctuations in time. However, this method should be adequate where the WL has been shown to be low, and fairly stable in time.

6.2 Area Continuous Sampling

This method solves the problem of not being able to measure large fluctuations of radon or thoron daughters but the difficulty of the results not being representative of the exposure to workers because of spatial variations remain. The equipment cost of this method of monitoring is considerable compared to grab sampling if more than a few monitoring stations are required. It is expected that this method would only be chosen over grab sampling in exceptional cases.

6.3 Active Radon (Thoron) Daughter Dosimeters

As mentioned above, these devices have received considerable attention, particularly in the last 10 years. They have the obvious advantage over area monitoring that the sampled air should be quite representative of the air inhaled by the wearer of the dosimeter. Recent developments have resulted in much more reliable pumping systems, and the weight and bulk can be kept acceptably small. The main disadvantage of these devices is cost, which of course increases with the number of units in service. It should also be appreciated that these dosimeters that are read out "after the fact" will not entirely replace area monitoring as area monitoring will still be required for exposure control purposes.

6.4 Semi-Passive Radon (Thoron) Daughter Dosimeters

These devices have the potential of being able to measure radon or thoron daughters at considerably less cost than an active device. However, there are serious difficulties in estimating and controlling their effective "pumping" rates. Because this "pumping" rate depends on the aerosol parameters, and on the relative humidity considerable effort would be required to demonstrate that these devices could meet the accuracy requirements suggested above.

6.5 Passive Radon (Thoron) Only Dosimeters

These dosimeters have the major disadvantage that they do not measure the hazard (radon or thoron daughters), which must be approximated from the estimated radon (thoron) concentration. If this disadvantage can be overcome, or at least if it can be shown that the uncertainty in estimating the WLM with these devices is within acceptable limits, they offer a method of estimating individual exposures directly within a reasonable cost. We feel that there is considerable promise in these devices, and that enough is known about the physical process involved in estimating radon (thoron) daughter concentrations from radon (thoron) concentrations that a study could be undertaken to determine the inherent uncertainties involved.

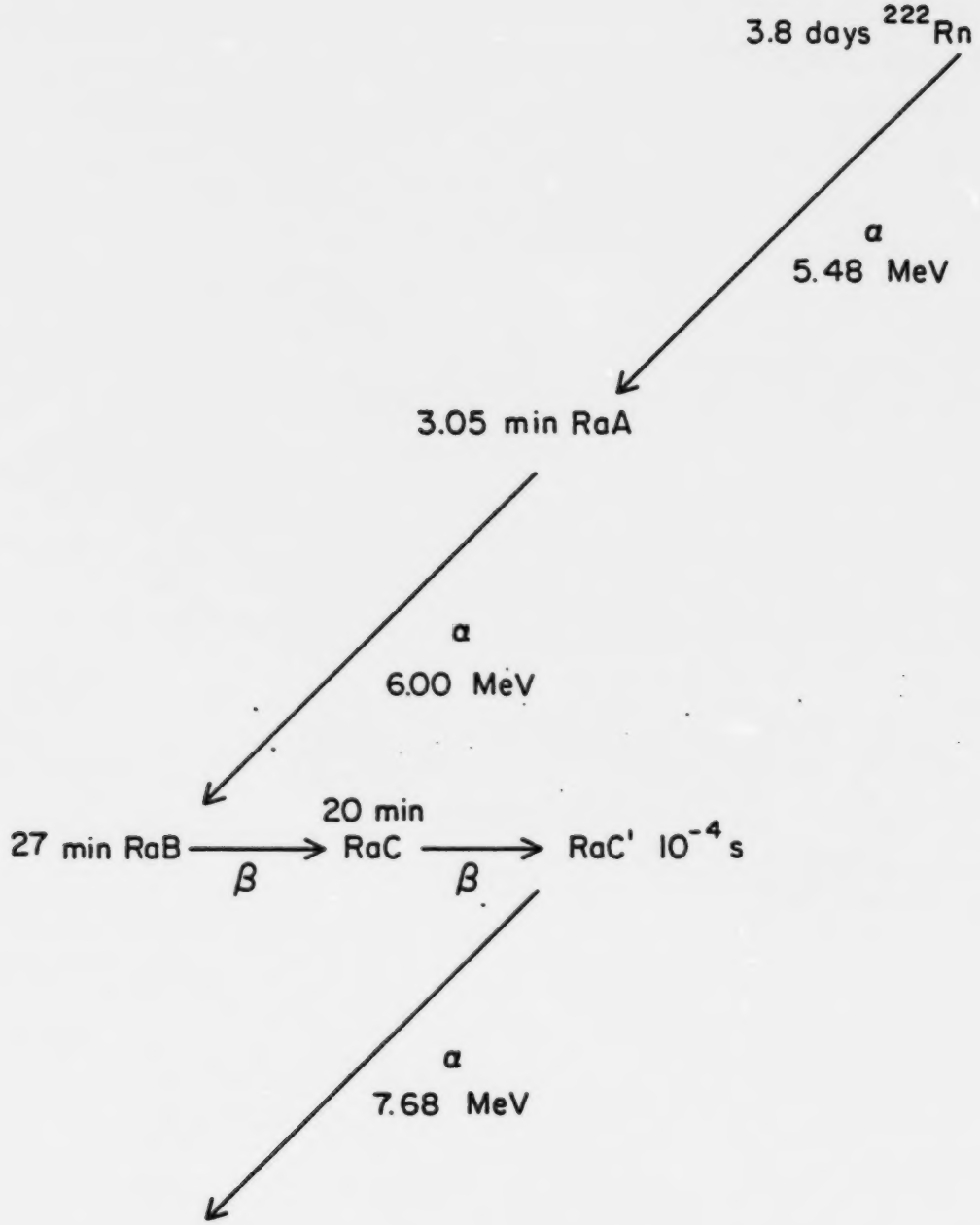

FIGURE 1 Part of a simplified version of the U-238 decay series showing Rn-222 and its short-lived daughters.

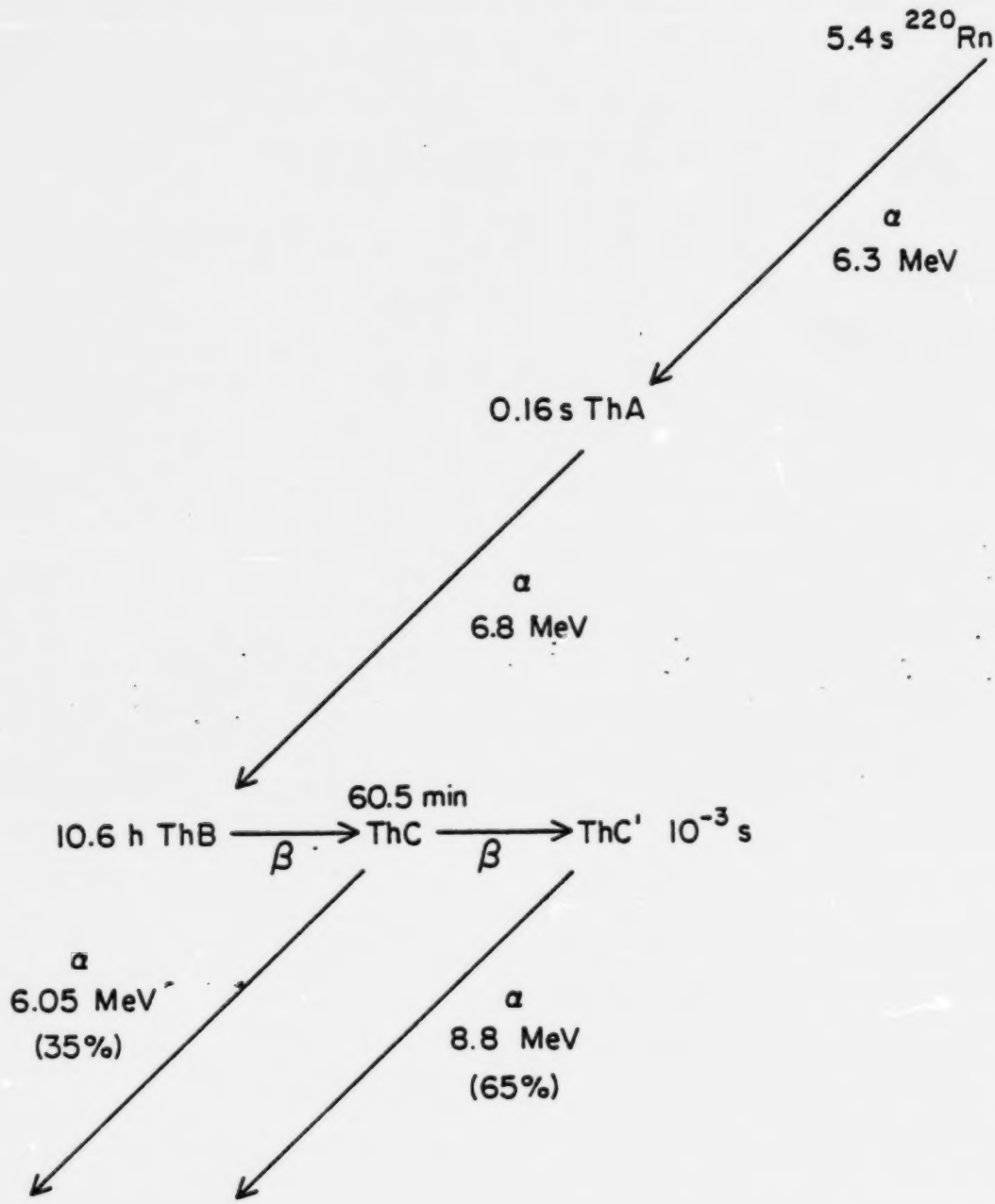

FIGURE 2 Part of a simplified version of the Th-232 decay series showing Rn-220 and its short-lived daughters.

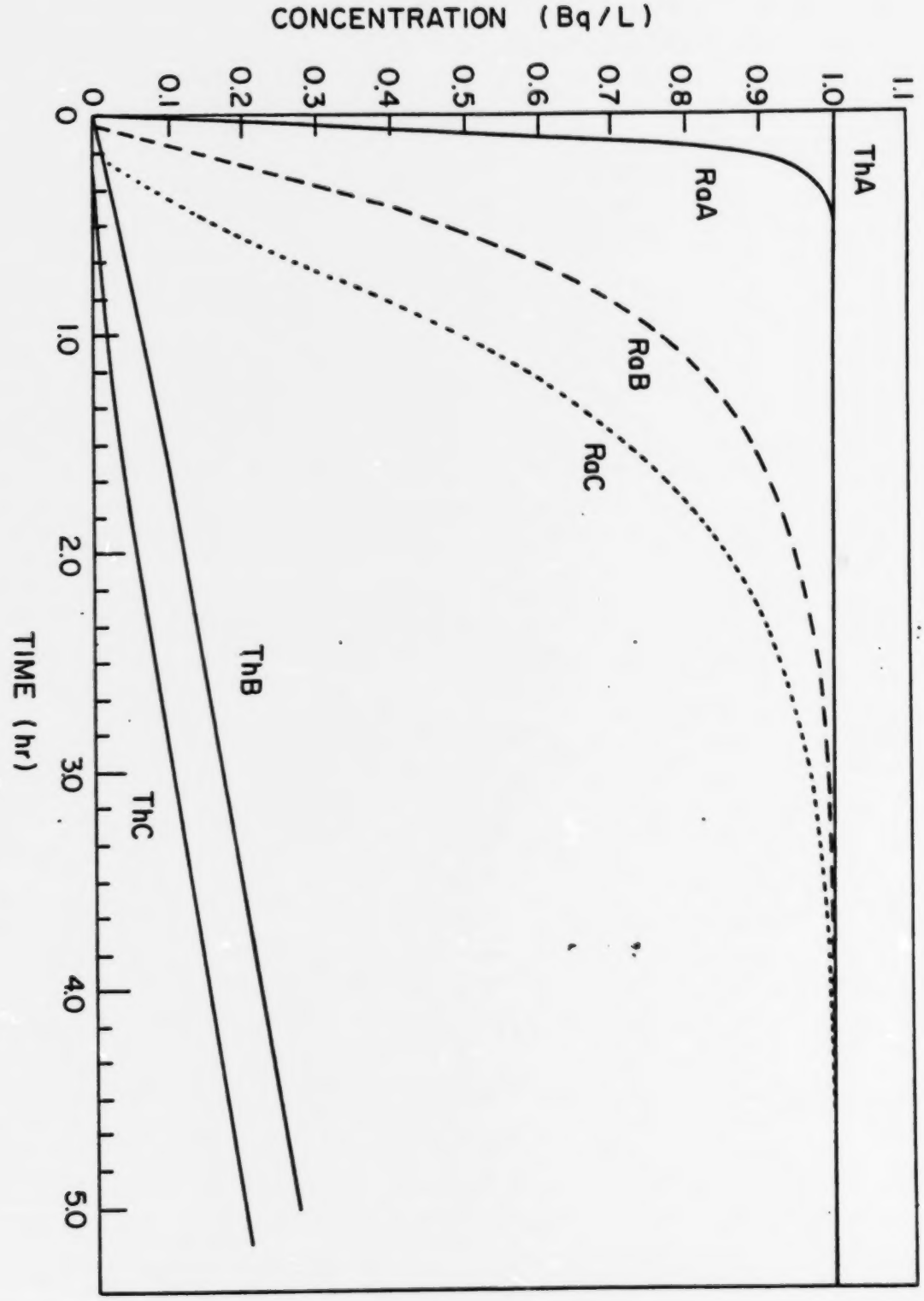

FIGURE 3 The build-up of the short-lived daughters of Rn-220 and Rn-222 as a function of time when the concentration of the parent is constant at 1 Bq/L.

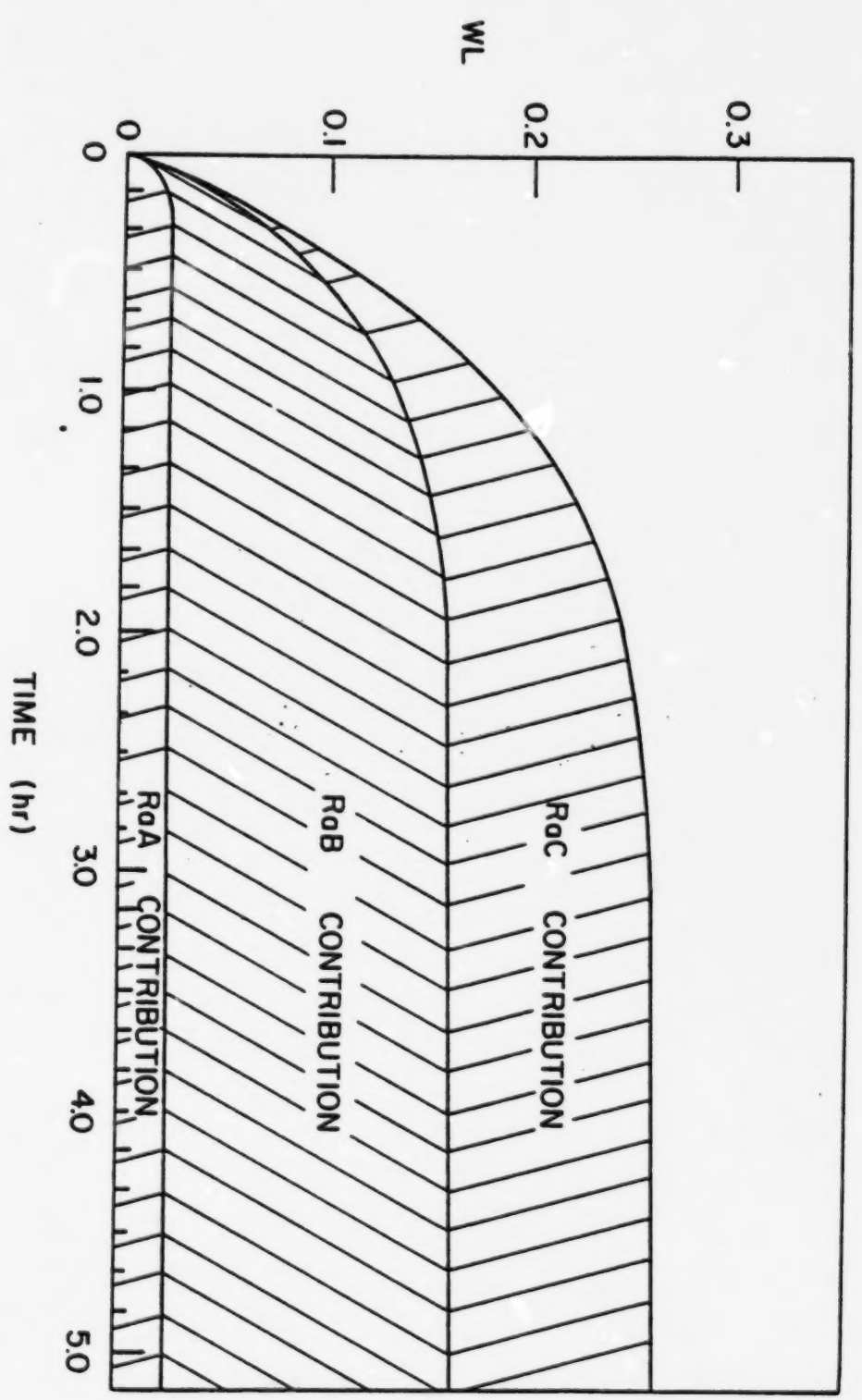

FIGURE 4 The build-up of the working level as a function of time for a constant Rn-222 concentration of 1 Bq/L. The contribution of each of the short-lived daughters is shown.

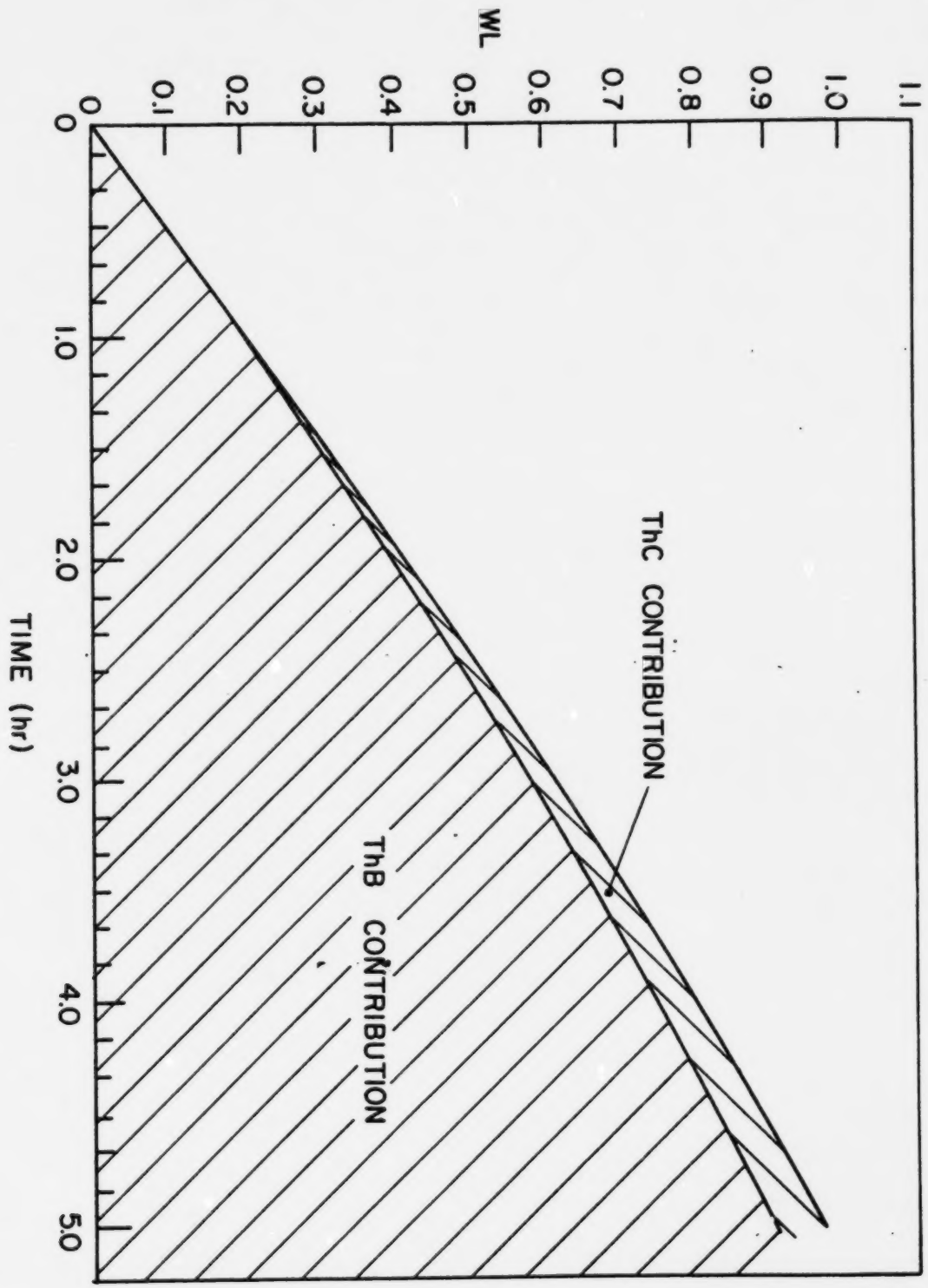

FIGURE 5 The build-up of the working level as a function of time for a constant Rn-220 concentration of 1 Bq/L. The individual contribution of ThC and ThB is shown. The contribution of ThA is too small to be shown on the figure.

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APPENDIX A

The Determination of WL Using Nuclear Track Detectors

In an active dosimeter, the airborne daughters are collected on a filter and the decays subsequently recorded. The total alpha energy is registered. The working level can thus be determined.

If a track detector is used, it is desireable to determine the alpha energy corresponding to each track. If the alpha energies are determined, then the working level is given by (for radon daughters)

$$WL = Na\ Ea + Nc'\ Ec'$$

Na number of tracks due to RaA
Ea alpha energy of RaA
Nc' number of tracks due to RaC'
Ec' alpha energy of RaC'

If only the total number of tracks, $N = Na + Nc'$, is measured, then the working level may be estimated by

$$WL \sim N\ E$$

where E is chosen such that $Ea < E < Ec'$.

The best choice for E will be dependent upon the age of the air being measured. If E is chosen to be, for example, (0.5 Ea + 0.5 Ec'), the maximum possible error from this source is less than 15%, regardless of the age of the air.

In a passive dosimeter, the situation is less favourable. Because the short-lived daughters are not deliberately trapped and held until they have decayed, the total alpha energy cannot be directly measured. All that can be directly detected are degraded alpha particles from the alpha active daughters in the air, as well as spurious alphas from plated out daughters and from the parent radon and thoron.

For the sake of the following discussion, we will assume that the track detector has sufficient resolution to allow the unfolding of the collected alpha spectrum to allow the separate determination of the concentration in air of the various species. The practical realization of this assumption is highly unlikely at the present time.

Now

$$WL(Rn) = (0\ C(Rn) + 2.4E\text{-}2\ C(RaA) + 1.4E\text{-}1\ C(RaB) + 1.0E\text{-}1\ C(RaC))\ (WL\ L/Bq)$$

$$WL(Tn) = (0\ C(Tn) + 2.6E\text{-}5\ C(ThA) + 3.3E{+}0\ C(ThB) + 3.1E\text{-}1\ C(ThC))\ (WL\ L/Bq)$$

C(...) is the concentration of the indicated species in $Bq\,L^{-1}$.

It is thus seen that the working levels are independent of some concentrations that we measure whether we want to or not (Rn and Tn) and strongly dependent on two concentrations that we can't measure even in principle (RaB and ThB). It is important to note that these two invisible concentrations are the single most important contributions to their respective working levels. The problem is particularly acute for thoron where C(ThB) is an order of magnitude more important than either C(ThA) or C(ThC). This situation is a consequence of the longer half lives of these two beta active daughters.

We are thus in the situation that the working level is not uniquely determined by the data available. We know however that C(RaA) > C(RaB) > C(RaC). Indeed, if the age of the air were a useful characterization then a knowledge of C(RaA) and C(RaC) would be all that was necessary to determine C(RaB). Figure 6 presents data to show that while the age of the air has some qualitative meaning, it cannot be used to make quantitative predictions. This is because any real sample of air is in fact a mixture of air of many different ages. The conclusion is that we can't uniquely determine C(RaB) from C(RaA) and C(RaC).

In summary, even if a passive dosimeter were capable of determining the concentration in air of all of the available alpha emitters present, the accuracy of the working level determination will be limited by the inability of the track detector to measure the most important contributer to the working level. Some progress can be made by estimating C(RaB) and C(ThB) from the concentration of the other daughter products, but the fundamental limitation remains.

We have not evaluated what is the best theoretical accuracy that could be obtained without a direct measure of the concentration of the beta emitting daughters, but approximations that limit the maximum possible error to less than 40% for radon daughters are readily derivable. The major difficulty encountered is what criteria to use to optimize the approximation.

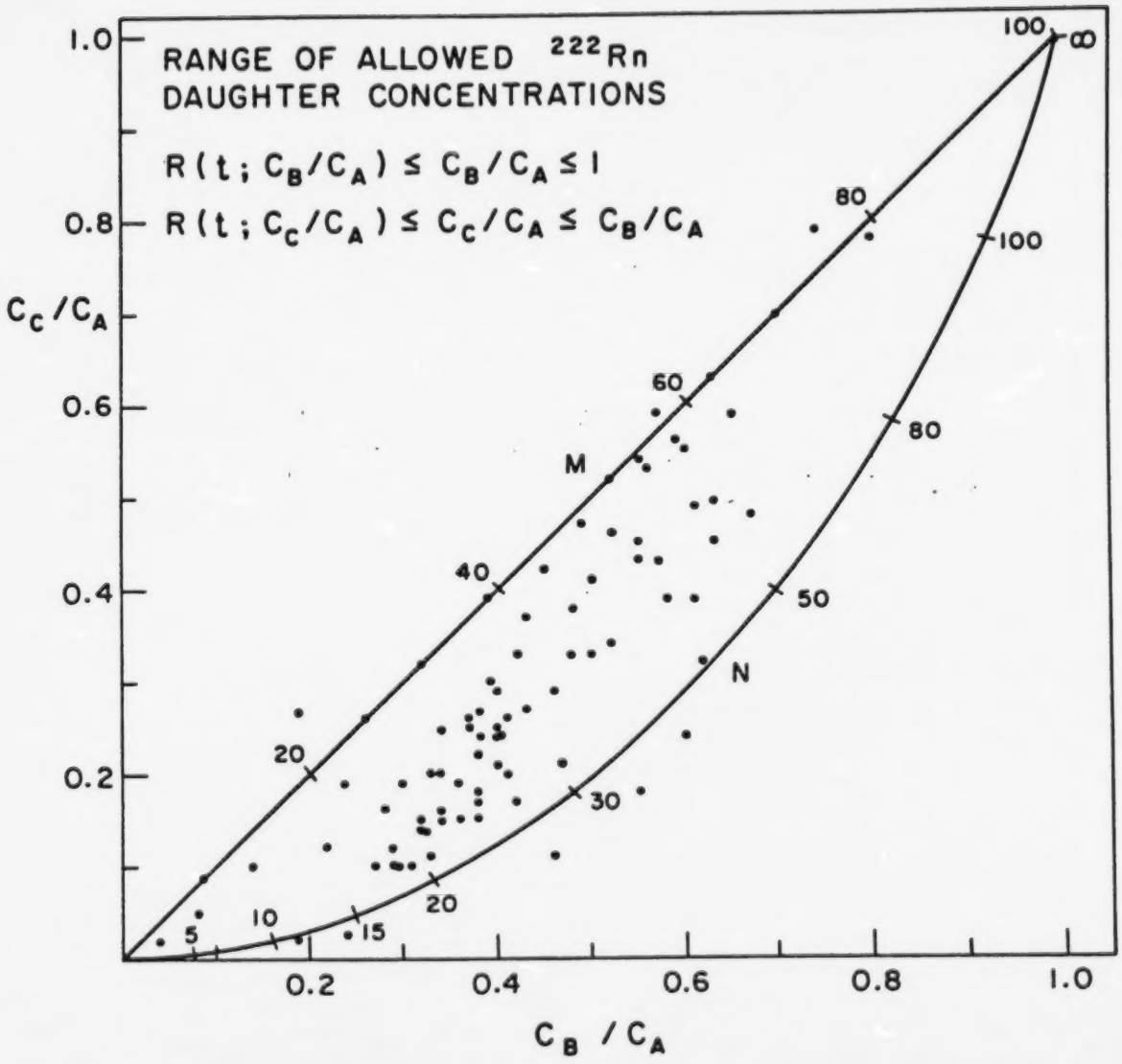

FIGURE 6 The range of Rn-222 daughter activity concentration ratio measurements in mines. Curve N represents young air as it ages whereas curve M is the limit of mixtures of young air (curve M) and very old air (adapted from Johnson [77]).

APPENDIX B

Literature Review

A computer search was made of the INIS (International Nuclear Information System) and INSPEC (Information Services for the Physics and Engineering Communities) data bases for articles on passive radon dosimeters and another search for radon dosimeters in general. The searches covered the years from 1974 to the present. The search did not explicitly mention radon daughters, but neither were they excluded. These searches produced a list of 93 references but with some duplication. This list was reduced by removing duplications, foreign language publications and all publications in summary form only.

Some of the references could be eliminated on the basis of their titles alone. This was mainly used to eliminate obviously spurious references. The remaining references were checked, if possible, to the extent of reading the published abstract in INIS ATOMINDEX. Because the search procedure did not differentiate between radon and radon daughters, there were some references which were relevant to radon dosimeters but with no direct connection to passive radon daughter dosimeters. Some of these references were maintained for comparison purposes, but not all. Similarly, some references pertaining to active dosimeters were also collected.

Copies were made of all potentially relevant references (except for large reports). If a reference was not available locally, it was ordered. The computer located references were supplemented by manually located references. These were mainly AECL or AECB reports or journal articles too recent to be included on the computer data bases.

The reference lists of all collected articles were checked for other useful references. Some additional useful references were found in this way. At one point it appeared that some references were missed in the original computer search by not including "working level" in the search criteria. A second search was made based on "working level" in combination with "dosimeter" and some other possibilities such as "measurement" etc. This search did not turn up additional useful references but did produce many spurious references.

We have reviewed well over 100 papers, articles and reports. These papers are concerned with passive radon daughter dosimeters and related fields. A complete evaluation of the problems and potential of passive radon daughter dosimeters must include some discussion of active dosimeters, TLDs, track detectors, electrets, aerosols etc. etc... All of these fields have an extensive literature of their own. It is not practical to identify all of the relevant references in these subsidiary fields and we have not attempted to do so. We do believe, however, that we have collected articles pertaining to all of the key developments in passive radon daughter dosimeters and most of the aspects of the various subfields.

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